

## PATENT ABSTRACTS OF JAPAN

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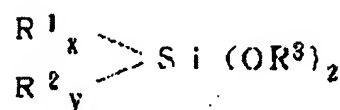
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## (54) PRODUCTION OF PROPYLENE COPOLYMER

## (57)Abstract:

PURPOSE: To obtain a propylene copolymer having well balanced transparency and rigidity by copolymerizing propylene with a small amount of ethylene using a specific polymerization catalyst component.

CONSTITUTION: This propylene copolymer having an ethylene content of 0.01-10wt. % can be produced by contacting (A) a solid catalyst component containing a metal oxide (preferably SiO<sub>2</sub>), Mg, Ti, a halogen and an electron- donative compound (e.g. di-n-butyl phthalate) as essential components with (B) an olefin in the presence of (C) an organic Al compound (preferably triethylaluminum) and (D) an organic silicon compound of the formula [R<sup>1</sup> in an N-containing heterocyclic substituent; R<sup>2</sup> is a 1-10C hydrocarbon group or R<sup>4</sup>O



(R<sup>4</sup> is a 3-10C hydrocarbon group); R<sup>3</sup> is CH<sub>3</sub> or C<sub>2</sub>H<sub>5</sub>; (x) is 1 or 2; (y) is 0 or 1; (z) is 2 or 3; x+y+z=4] (e.g. 2,3,4-trimethyl-3- azacyclopentyltrimethoxysilane) to obtain a polymerization catalyst component and copolymerizing propylene and ethylene in the presence of this catalyst component.

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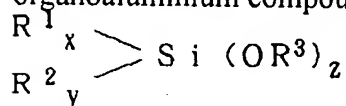
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 CLAIMS
 

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[Claim(s)]

[Claim 1] (A) the solid-state catalyst component which uses a metallic oxide, magnesium, titanium, a halogen, and an electron-donative compound as an indispensable component -- the (B) organoaluminium compound and the (C) general formula [\*\* 1]



[ -- R1 [ however, ] -- a nitrogen atom content heterocycle type substituent and R2 -- the hydrocarbon group of 1-10 carbon numbers -- R4 O-, R5 3 Si- or R6 3 SiO-, and R3 A methyl group or an ethyl group, x -- 1 or 2, and y -- 0 or 1, and z -- 2 or 3, and x+y+z=4 -- being shown -- R4 The hydrocarbon group of 3-10 carbon numbers, and R5 And R6 The hydrocarbon group of 1-10 carbon numbers is shown. ] The manufacture approach of the propylene copolymer of 0.01 - 10 % of the weight of ethylene contents which consist of copolymerizing a propylene and ethylene under existence of the polymerization catalyst component which comes out and is made to come to contact the (D) olefin under the existence of an organosilicon compound expressed.

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[Translation done.]

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DETAILED DESCRIPTION

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## [Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to the approach of manufacturing the polypropylene copolymer excellent in transparency and rigidity, in more detail about the manufacture approach of a propylene copolymer by carrying out random copolymerization of a propylene and the little ethylene using a specific catalyst component.

[0002]

[Description of the Prior Art] Although the high crystallinity polypropylene manufactured using the high stereoregularity catalyst of various classes is excellent in rigidity or thermal resistance, it is resin of opalescence and higher transparency may be desired depending on an application. As a means to improve transparency, in case the polymerization of the propylene is carried out, the method of adding little ethylene and manufacturing a random copolymer is learned well. However, although transparency improves the more the more there are many rates of the amount of ethylene to add, the crystallinity of polypropylene will be disturbed, as a result the rigid height which is the property of polypropylene original will be spoiled.

[0003] On the other hand, although various methods of improving rigidity are also proposed, those many are the approaches of performing after treatment of adding a nucleating additive etc. in polypropylene. Therefore, when becoming cost quantity from a process side, there are some which spoil the appearance of mold goods depending on an additive. The rigidity of all is insufficient although the method of improving rigidity with a polymerization method is also proposed without processing by the additive.

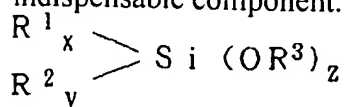
[0004]

[Problem(s) to be Solved by the Invention] This invention aims at offering the approach of obtaining the propylene copolymer excellent in the balance of transparency and rigidity.

[0005]

[Means for Solving the Problem] this invention persons completed a header and this invention for the ability of the purpose of this invention to be attained by carrying out the polymerization of the propylene with little ethylene using the specific polymerization catalyst component, as a result of inquiring wholeheartedly.

[0006] The summary of invention, i.e., the summary of this invention, is the (B) organoaluminium compound and the (C) general formula [\*\* 2] about the solid-state catalyst component which uses the (A) metallic oxide, magnesium, titanium, a halogen, and an electron-donative compound as an indispensable component.



[ -- R1 [ however, ] -- a nitrogen atom content heterocycle type substituent and R2 -- the hydrocarbon group of 1-10 carbon numbers -- R4 O-, R5 3 Si- or R6 3 SiO-, and R3 A methyl group or an ethyl

group,  $x = 1$  or  $2$ , and  $y = 0$  or  $1$ , and  $z = 2$  or  $3$ , and  $x+y+z=4$  -- being shown -- R4 The hydrocarbon group of 3-10 carbon numbers, and R5 And R6 The hydrocarbon group of 1-10 carbon numbers is shown. ] It comes out and the ethylene content which consists of copolymerizing a propylene and ethylene is in the manufacturing method of the propylene copolymer which is 0.01 - 10 % of the weight under existence of the polymerization catalyst component made to come to contact the (D) olefin under the existence of an organosilicon compound expressed.

[0007] Although the solid-state catalyst component (henceforth Component A) which is 1 of the catalyst component (henceforth the catalyst component of this invention) used by solid-state catalyst component this invention component uses a metallic oxide, magnesium, titanium, a halogen, and an electron-donative compound as an indispensable component It is prepared when such a component usually contacts a halogen content compound, respectively in the case of a metallic oxide, a magnesium compound, a titanium compound and an electron-donative compound, and the compound with which said each compound does not have a halogen further.

[0008] (1) the metallic oxide used by metallic-oxide this invention -- the [ of an element / periodic-table ] -- the [ II group - ] -- if it is the oxide of the element chosen from the group of IV group's element and they are illustrated -- B-2 O<sub>3</sub>, MgO, aluminum 2O<sub>3</sub>, SiO<sub>2</sub>, CaO, TiO<sub>2</sub>, ZnO, ZrO<sub>2</sub>, SnO<sub>2</sub>, BaO, and ThO<sub>2</sub> etc. -- it is mentioned. They are B-2 O<sub>3</sub>, MgO, aluminum 2O<sub>3</sub>, SiO<sub>2</sub>, TiO<sub>2</sub>, and ZrO<sub>2</sub> also in these. It is desirable and is especially SiO<sub>2</sub>. It is desirable. Furthermore, the multiple oxide containing these metallic oxides, for example, SiO<sub>2</sub>-MgO, SiO<sub>2</sub>-aluminum 2O<sub>3</sub>, SiO<sub>2</sub>-TiO<sub>2</sub>, SiO<sub>2</sub>-V<sub>2</sub>O<sub>5</sub>, SiO<sub>2</sub>-Cr 2O<sub>3</sub>, SiO<sub>2</sub>-TiO<sub>2</sub>-MgO, etc. can be used. As for the configuration of these metallic oxides, a powder-like thing is usually used. Since forms, such as powdered magnitude and a configuration, affect the form of the olefin polymer obtained in many cases, adjusting suitably is desirable. As for a metallic oxide, it is desirable to deal with it so that it may calcinate at an elevated temperature as much as possible and may not contact atmospheric air and directly further from the purpose which removes the poisoning matter in use.

[0009] (2) A magnesium compound magnesium compound is a general formula MgR seven R8. It is expressed. It sets at a ceremony and is R7 and R8. Whether to be the same, and a different hydrocarbon group and OR9 A radical (R9 is a hydrocarbon group) and a halogen atom are shown. In a detail, it is R7 more. And  $R < \sup > 8$  As a hydrocarbon group, the alkyl group of 1-20 carbon numbers, a cycloalkyl radical, an aryl group, and an aralkyl radical are R9 as nine OR. The alkyl group of 1-12 carbon numbers, a cycloalkyl radical, an aryl group, and an aralkyl radical are mentioned for chlorine, a bromine, iodine, fluorine, etc. as a halogen atom.

[0010] Although the example of these compounds is shown below, it sets in a chemical formula, and they are Me:methyl, Et:ethyl, and Pr.: Propyl, Bu:butyl, helium:hexyl, Oct:octyl, Ph:phenyl, cyHe:Cyclohexyl is shown, respectively. MgMe<sub>2</sub>, MgEt<sub>2</sub>, MgPr<sub>2</sub>, MgBu<sub>2</sub>, MgHe<sub>2</sub>, MgOct<sub>2</sub>, MgEtBu, MgPh<sub>2</sub>, MgcyHe<sub>2</sub>, Mg(OMe)<sub>2</sub>, Mg(OEt)<sub>2</sub>, Mg(Obu)<sub>2</sub>, Mg(OHe)<sub>2</sub>, Mg(OOct)<sub>2</sub>, Mg(OPh)<sub>2</sub>, Mg(OcyHe)<sub>2</sub>, EtMgCl, BuMgCl, HeMgCl, i-BuMgCl, t-BuMgCl, PhMgCl, PhCH<sub>2</sub> MgCl, EtMgBr, BuMgBr, PhMgBr, BuMgI, EtOMgCl, BuOMgCl, HeOMgCl, PhOMgCl, EtOMgBr, BuOMgBr, EtOMgI, MgCl<sub>2</sub>, MgBr<sub>2</sub>, and MgI<sub>2</sub>.

[0011] In case the above-mentioned magnesium compound prepares Component A, it can also be prepared from metal magnesium or other magnesium compounds. As the example, they are metal magnesium, halogenated hydrocarbon, and a general formula.  $X_n M (OR)^{m-n}$  In an alkoxy group content compound [type, in X, boron, carbon, aluminum, silicon or a phosphorus atom, and R show the hydrocarbon group of 1-20 carbon numbers, and, as for a hydrogen atom, a halogen atom or the hydrocarbon group of 1-20 carbon numbers, and M, m shows the valence of M, and  $m > n \geq 0$ . ] The method of making it contact is mentioned. As a hydrocarbon group of X and R of the general formula of this alkoxy group content compound Methyl (Me) and ethyl (Et) Propyl, (Pr), and i-propyl (i-Pr), butyl (Bu) i-butyl (i-Bu) and hexyl (helium) Alkyl groups, such as octyl (Oct), and cyclohexyl (cyHe), Alkenyl radicals, such as cycloalkyl radicals, such as methylcyclohexyl, an allyl compound, propenyl, and butenyl, and phenyl (Ph) Aralkyl radicals, such as aryl groups, such as tolyl and xylyl, phenethyl, and 3-phenylpropyl, etc. are mentioned. Also in these, especially the alkyl group of 1-10 carbon

numbers is desirable. Hereafter, the example of an alkoxy group content compound is given.

[0012] \*\* Compound type in case M is carbon C (OR)<sub>4</sub> C(OMe)<sub>4</sub> boiled and contained and C (OEt)<sub>4</sub>, C(OPr)<sub>4</sub> and C (OBu)<sub>4</sub>, C(Oi-Bu)<sub>4</sub> and C (OHe)<sub>4</sub>, C(OOct)<sub>4</sub>: Formula XC (OR)<sub>3</sub> HC (OMe)<sub>3</sub> boiled and contained, HC (OEt)<sub>3</sub>, HC (OPr)<sub>3</sub>, HC (OBu)<sub>3</sub>, HC (OHe)<sub>3</sub>, HC<sub>3</sub> (OPh) ; MeC (OMe)<sub>3</sub>, MeC (OEt)<sub>3</sub>, and EtC (OMe)<sub>3</sub> , EtC (OEt)<sub>3</sub>, and cyHeC (OEt)<sub>3</sub>, PhC (OMe)<sub>3</sub>, PhC (OEt)<sub>3</sub>, CH<sub>2</sub>ClC (OEt)<sub>3</sub>, MeCHBrC (OEt)<sub>3</sub>, MeCHClC<sub>3</sub> (OEt) ; ClC (OMe)<sub>3</sub>, ClC (OEt)<sub>3</sub>, ClC (Oi-Bu)<sub>3</sub>, and BrC (OEt)<sub>3</sub> ; Formula X<sub>2</sub> C<sub>2</sub> (OR) MeCH (OMe)<sub>2</sub> contained, MeCH (OEt)<sub>2</sub>, CH<sub>2</sub> 2 (OMe), CH<sub>2</sub> 2 (OEt), CH<sub>2</sub> ClCH (OEt)<sub>2</sub>, CHCl<sub>2</sub> CH (OEt)<sub>2</sub>, and CCl<sub>3</sub> CH (OEt)<sub>2</sub>, CH<sub>2</sub> BrCH (OEt)<sub>2</sub>, and PhCH<sub>2</sub> (OEt).

[0013] \*\* Compound type in case M is silicon Si (OR)<sub>4</sub> Si boiled and contained 4 Si (OMe) 4 Si (OEt) (OBu) 4, Si (Oi-Bu)<sub>4</sub>, Si (OHe)<sub>4</sub>, Si (OOct)<sub>4</sub>, Si<sub>4</sub> (OPh) : Formula XSi<sub>3</sub> (OR) HSi (OEt)<sub>3</sub> contained, HSi (OBu)<sub>3</sub>, HSi (OHe)<sub>3</sub>, and HSi (OPh)<sub>3</sub> ; MeSi 3 MeSi (OMe) (OEt) 3, MeSi (OBu)<sub>3</sub>, EtSi (OEt)<sub>3</sub>, PhSi (OEt)<sub>3</sub>, EtSi<sub>3</sub> (OPh) ; ClSi (OMe)<sub>3</sub>, ClSi (OEt)<sub>3</sub>, ClSi (OBu)<sub>3</sub>, ClSi (OPh)<sub>3</sub>, and BrSi (OEt)<sub>3</sub> ; Formula X<sub>2</sub>Si<sub>2</sub> (OR) Me<sub>2</sub> contained Si (OMe)<sub>2</sub>, Me<sub>2</sub>Si (OEt)<sub>2</sub>, and Et<sub>2</sub> Si<sub>2</sub> (OEt) ; MeClSi (OEt)<sub>2</sub> ; CHCl<sub>2</sub> SiH<sub>2</sub> (OEt) ; CCl<sub>3</sub> SiH<sub>2</sub> (OEt) ; MeBuSi<sub>2</sub> (OEt) : Me<sub>3</sub> SiOMe contained in X<sub>3</sub> SiOR, Me<sub>3</sub> SiOEt, Me<sub>3</sub>SiOBu, Me<sub>3</sub> SiOPh, Et<sub>3</sub> SiOEt, Ph<sub>3</sub> SiOEt.

[0014] \*\* Compound type in case M is boron B (OR) 3 B(OEt) 3 contained, B(OBu) 3, B(OHe) 3, and B(OPh) 3.

[0015] \*\* Compound type in case M is aluminum aluminum<sub>3</sub> (OR) aluminum (OMe)<sub>3</sub> contained, aluminum (OEt)<sub>3</sub>, aluminum (OPr)<sub>3</sub>, aluminum (Oi-Pr)<sub>3</sub>, aluminum (OBu)<sub>3</sub>, aluminum (Ot-Bu)<sub>3</sub> and aluminum (OHe)<sub>3</sub>, and aluminum<sub>3</sub> (OPh).

[0016] \*\* Compound type in case M is phosphorus P (OR) 3 P(OMe) 3 contained, P(OEt) 3, P(OBu) 3, P(OHe) 3, and P(OPh) 3.

[0017] furthermore, said magnesium compound -- the [ periodic-table ] -- II group or \*\* A complex with the organic compound of an IIIa group metal (M') can also be used. This complex is a general formula. MgR seven R<sub>8</sub> and n<sub>0</sub> (M'R<sub>10m0</sub>) It is expressed. As this metal, it is aluminum, zinc, calcium, etc. and R<sub>10</sub> is the alkyl group of 1-12 carbon numbers, a cycloalkyl radical, an aryl group, and an aralkyl radical. Moreover, m<sub>0</sub> It is the valence of metal M' n<sub>0</sub> The number of 0.1-10 is shown. as the example of a compound expressed with M'R<sub>10m0</sub> -- AlMe<sub>3</sub>, AlEt<sub>3</sub>, Ali-Bu<sub>3</sub>, AlPh<sub>3</sub>, ZnMe<sub>2</sub>, ZnEt<sub>2</sub>, ZnBu<sub>2</sub>, ZnPh<sub>2</sub>, CaEt<sub>2</sub>, and CaPh<sub>2</sub> etc. -- it is mentioned.

[0018] (2) if a titanium compound titanium compound is a compound of bivalence and trivalent and tetravalent titanium and they are illustrated -- a titanium tetrachloride -- titanium, TORIKURORU ethoxy titanium, TORIKURORU butoxytitanium, dichloro diethoxy titanium, dichlor dibutoxy titanium, JIKURORUJI phenoxy titanium, KURORU triethoxytitanium, KURORU tributoxytitanium, tetrabutoxytitanium, a titanium trichloride, etc. can be mentioned 4 bromination. Also in these, tetravalent titanium halogenides, such as a titanium tetrachloride, TORIKURORU ethoxy titanium, dichlor dibutoxy titanium, and JIKURORUJI phenoxy titanium, are desirable, and especially a titanium tetrachloride is desirable.

[0019] (3) The phosphorus combined as an electron-donative compound electron-donative compound through carboxylic acids, a carboxylic anhydride, carboxylate, a carboxylic-acid halogenide, alcohols, ether, ketones, amines, amides, nitril, aldehydes, alcoholate, an organic radical, carbon, or oxygen, an arsenic and an antimony compound, phosphoamides, thioether, thioester, a carbonate, etc. are mentioned. Carboxylic acids, a carboxylic anhydride, carboxylate, a carboxylic-acid halogenide, alcohols, and ether are preferably used among this.

[0020] As an example of a carboxylic acid, a formic acid, an acetic acid, a propionic acid, butanoic acid, an isobutyric acid, Aliphatic series monocarboxylic acid, such as a valeric acid, a caproic acid, pivalate, an acrylic acid, a methacrylic acid, and a crotonic acid, A malonic acid, a succinic acid, a glutaric acid, an adipic acid, a sebacic acid, a maleic acid, Aliphatic series hydroxy acid, such as aliphatic series dicarboxylic acid, such as a fumaric acid, and a tartaric acid, Cyclohexane monocarboxylic acid, cyclohexene monocarboxylic acid, cis- - 1, 2-cyclohexane dicarboxylic acid, Alicyclic carboxylic acids, such as the cis--4-methyl cyclohexene -1 and 2-dicarboxylic acid, A benzoic acid, a toluic acid, an anisic acid, p-tertiary butyl benzoic acid, a naphthoic acid, Aromatic series multiple-valued carboxylic acids,

such as aromatic series monocarboxylic acid, such as a cinnamic acid, a phthalic acid, isophthalic acid, a terephthalic acid, naphthalic acid, trimellitic acid, hemimellitic acid, trimesic acid, pyromellitic acid, and mellitic acid, etc. are mentioned. As a carboxylic anhydride, the acid anhydride of the above-mentioned carboxylic acids can use it.

[0021] As carboxylate, the above-mentioned monochrome or the multiple-valued ester of carboxylic acids can be used. As the example Formic-acid butyl, ethyl acetate, butyl acetate, isobutyl isobutyrate, pivalate propyl, Pivalate isobutyl, an ethyl acrylate, a methyl methacrylate, ethyl methacrylate, Methacrylic-acid isobutyl, a diethyl malonate, malonic-acid diisobutyl, Diethyl succinate, succinic-acid dibutyl, succinic-acid diisobutyl, glutaric-acid diethyl, Glutaric-acid dibutyl, glutaric-acid diisobutyl, diisobutyl adipate, A dibutyl sebacate, sebacic-acid diisobutyl, a diethyl maleate, Dibutyl maleate, maleic-acid diisobutyl, fumaric-acid monomethyl, Diethyl fumarate, diisobutyl fumarate, tartaric-acid diethyl, dibutyl tartrate, Tartaric-acid diisobutyl, cyclohexane-carboxylic-acid ethyl, methyl benzoate, Ethyl benzoate, para-toluylic-acid methyl, p-tertiary butyl ethyl benzoate, p-anisic-acid ethyl, alpha-naphthoic-acid ethyl, alpha-naphthoic-acid isobutyl, Ethyl cinnamate, phthalic-acid monomethyl, phthalic-acid monobutyl, dibutyl phthalate, Diisobutyl phthalate, phthalic-acid dihexyl, a dioctyl phthalate, Di-2-ethylhexyl phthalate, diallyl phthalate, diphenyl phthalate, Diethyl isophthalate, diisobutyl isophthalate, diethyl terephthalate, Terephthalic-acid dibutyl, naphthalic acid diethyl, naphthalic acid dibutyl, trimellitic acid triethyl, trimellitic acid tributyl, pyromellitic acid tetramethyl, pyromellitic acid tetraethyl, pyromellitic acid tetrabutyl, etc. are mentioned.

[0022] As a carboxylic-acid halogenide, the acid halide of the above-mentioned carboxylic acids can be used. As the example Acetic-acid chloride, acetic-acid bromide, acetic-acid iodide, propionic-acid chloride, Butanoic acid chloride, butanoic acid bromide, butanoic acid iodide, pivalate chloride, Pivalate bromide, acrylic-acid chloride, acrylic-acid bromide, acrylic-acid iodide, Methacrylic-acid chloride, methacrylic-acid bromide, methacrylic-acid iodide, Crotonic-acid chloride, malonic-acid chloride, malonic-acid bromide, succinic-acid chloride, Succinic-acid bromide, glutaric-acid chloride, glutaric-acid bromide, adipic-acid chloride, Adipic-acid bromide, sebacic-acid chloride, sebacic-acid bromide, maleic-acid chloride, Maleic-acid bromide, fumaric-acid chloride, fumaric-acid bromide, tartaric-acid chloride, Tartaric-acid bromide, cyclohexane-carboxylic-acid chloride, cyclohexane-carboxylic-acid bromide, 1-cyclohexene carboxylic-acid chloride, cis--4-methyl cyclohexene carboxylic-acid chloride, Cis--4-methyl cyclohexene carboxylic-acid bromide, a benzoyl chloride, bromination -- benzoyl, para-toluylic-acid chloride, and para-toluylic-acid bromide -- p-anisic-acid chloride, p-anisic-acid bromide, alpha-naphthoic-acid chloride, cinnamic-acid chloride, cinnamic-acid bromide, phthalic-acid dichloride, and a phthalic acid -- a jib -- ROMIDO, isophthalic acid dichloride, and isophthalic acid -- a jib -- ROMIDO, terephthalic acid dichloride, and naphthalic acid dichloride are mentioned. Moreover, the monoalkyl halogenide of dicarboxylic acid like adipic-acid monomethyl chloride, maleic-acid monoethyl chloride, maleic-acid monomethyl chloride, and phthalic-acid butyl chloride can also be used.

[0023] Alcohols are general formulas. It is expressed with R11OH. In a formula, R11 is the alkyl of 1-12 carbon numbers, the alkenyl, cycloalkyl, aryl, and the aralkyl. As the example, they are a methanol, ethanol, propanol, isopropanol, a butanol, isobutanol, a pentanol, a hexanol, an octanol, 2-ethylhexanol, a cyclohexanol, benzyl alcohol, allyl alcohol, a phenol, cresol, a xylenol, ethylphenol, an isopropyl phenol, p-tertiary butylphenol, n-octyl phenol, etc.

[0024] Ether is expressed with general formula R12OR13. in a formula, R12 and R13 are the alkyl of 1-12 carbon numbers, the alkenyl, cycloalkyl, aryl, and the aralkyl, and R13 may be the same as R12, or may differ. As the example, they are diethylether, diisopropyl ether, dibutyl ether, the diisobutyl ether, JiISO amyl ether, the G 2-ethylhexyl ether, diaryl ether, the ethyl allyl compound ether, the butyl allyl compound ether, diphenyl ether, an anisole, ethyl phenyl ether, etc.

[0025] As the method of preparation of Component A, \*\* metallic oxide (component 1), a magnesium compound (component 2), a titanium compound (component 3), and an electron-donative compound (component 4) are contacted in the sequence. \*\* Contact a component 4 and a component 3 in the sequence after contacting a component 1 and a component 2. \*\* After contacting a component 1 and a

component 2, use and contact a component 3 and a component 4 to coincidence. \*\* After contacting to coincidence the \*\* component 2, the component 3, and component 4 which contact a component 3 and a component 1 in the sequence after contacting the \*\* component 2 and component 4 which contact a component 4 and a component 1 in the sequence after contacting a component 2 and a component 3, the approach of contacting a component 1 can adopt. Moreover, before making it contact using a component 3, a halogen content compound can also be made to contact.

[0026] As a halogen content compound, they are halogenated hydrocarbon, halogen content alcohol, the silicon halide compound that has hydrogen-silicon association, and \*\*\*\*\*. The halogenide (henceforth metal halide) of an IIIa group, an IVa group, and Va group element etc. is mentioned.

[0027] As halogenated hydrocarbon, it is the monochrome and the Pori halogenation object of the aliphatic series of the saturation of 1-12 carbon numbers, or partial saturation, alicyclic, and aromatic hydrocarbon. The concrete example of these compounds in an aliphatic compound Methyl chloride, a methyl bromide, methyl iodide, a methylene chloride, A methylene star's picture, methylene iodide, chloroform, bromoform, Iodoform, a carbon tetrachloride, carbon tetrabromide, carbon tetraiodide, ethyl chloride, An ethyl star's picture, ethyl iodide, 1,2-dichloroethane, 1, 2-dibromoethane, 1, 2-diiodoethane, methyl chloroform, Methyl bromoform, methyl iodoform, 1 and 1, 2-trichloroethylene, 1, 1, 2-TORIBUROMO ethylene, 1, 1 and 2, 2-tetrachlorethylene, Pentachloroethane, hexa chloroethane, hexa bromoethane, n-propyl chloride, 1, a 2-dichloro propane, hexachloropropylene, OKUTA chloropropane, Deca BUROMO butane and chlorinated paraffin in an alicyclic compound A chloro cyclopropane, A tetra-KURORU cyclopentane, hexachlorocyclopentadiene, and a hexa KURORU cyclohexane In an aromatic compound, chlorobenzene, a bromobenzene, o-dichlorobenzene, p-dichlorobenzene, hexachlorobenzene, hexabromobenzene, benzotrichloride, p-chlorobenzo trichloride, etc. are mentioned. Two or more sorts of these compounds may be used only not only in a kind.

[0028] The compound with which the piece of arbitration other than a hydroxyl group in the monochrome which has a piece or two or more hydroxyl groups, or polyhydric alcohol, or two or more hydrogen atoms were permuted by the halogen atom in the monad as halogen content alcohol is meant. As a halogen atom, although chlorine, a bromine, iodine, and a fluorine atom are mentioned, a chlorine atom is desirable. When these compounds are illustrated, 2-chloroethanol, 1-Krol-2-propanol, 3-Krol-1-propanol, 1-Krol-2-methyl-2-propanol, 4-Krol-1-butanol, 5-Krol-1-pentanol, a 6-Krol-1-hexanol, 3-Krol-1, 2-propanediol, 2-KURORU cyclohexanol, 4-KURORU benzhydrol, (m, o, p)-KURORU benzyl alcohol, 4-KURORU catechol, 4-Krol -(m, o)- Cresol, 6-Krol -(m, o)- Cresol, 4-Krol -3, 5-dimethylphenol, KURORU hydroquinone, A 2-benzyl-4-KURORU phenol, a 4-Krol-1-naphthol, (m, o, p)-KURORU phenol, p-Krol-alpha methylbenzyl alcohol, 2-Krol-4-phenylphenol, 6-KURORU thymol, 4-KURORU resorcinol, 2-bromine ethanol, 3-bromine-1-propanol, 1-bromine-2-propanol, 1-bromine-2-butanol, 2-bromine-p-cresol, 1-bromine-2-naphthol, 6-bromine-2-naphthol, (m, o, p)-bromine phenol, 4-bromine resorcinol, (m, o, p)-FURORO phenol, p-iodo phenol : 2 and 2-dichloro ethanol, 2, 3-dichloro-1-propanol, 1, 3-dichloro-2-propanol, 3-Krol-1-(alpha-KURORU methyl)-1-propanol, 2, and 3-a jib -- ROM-1-propanol -- 1 and 3- a jib -- ROM-2-propanol, 2, and 4- a jib -- a ROM phenol -- 2 and 4- a jib -- ROM-1-naphthol: -- 2, 2, and 2-trichloroethanol -- A 1, 1, and 1-Tori Krol-2-propanol, beta and beta, and beta-Tori Krol-tert-butanol, 2, 3, 4-TORIKURORU phenol, 2 and 4, 5-TORIKURORU phenol, 2, 4, 6-TORIKURORU phenol, 2 and 4, 6-tribromophenol, 2, 3, 5-TORIBUROMU-2-hydroxytoluene, 2 and 3, 5-TORIBUROMU-4-hydroxytoluene, 2, 2, and 2-trifluoro ethanol, alpha and alpha, and alpha-trifluoro-m-cresol, 2, 4, 6-TORIIODO phenol : 2, 3, 4, a 6-tetra-KURORU phenol, Tetra-KURORU hydroquinone, tetra-KURORU bisphenol A, tetra-bromine bisphenol A, 2, 2 and 3, 3-tetrafluoro-1-propanol, 2, 3 and 5, 6-tetrafluoro phenol, tetrafluoro resorcinol, etc. are mentioned.

[0029] As a silicon halide compound which has hydrogen-silicon association  $\text{HSiCl}_3$ ,  $\text{H}_2\text{SiCl}_2$ , and  $\text{H}_3\text{SiCl}$ ,  $\text{HCH}_3\text{SiCl}_2$ , and  $\text{HC}_2\text{H}_5\text{SiCl}_2$ ,  $\text{H}(\text{t-C four H}_9)\text{SiCl}_2$ ,  $\text{HC}_6\text{H}_5\text{SiCl}_2$ ,  $\text{H}(\text{CH}_3)_2\text{SiCl}$ ,  $\text{H}(\text{i-C three H}_7)_2\text{SiCl}$ ,  $\text{H}_2\text{C}_2\text{H}_5\text{SiCl}$ ,  $\text{H}_2\text{SiCl}(\text{n-C four H}_9)$ ,  $\text{H}_2\text{SiCl}(\text{C}_6\text{H}_4\text{CH}_3)$  and  $\text{HSiCl}(\text{C six H}_5)_2$  etc. -- it is mentioned.

[0030] As metal halide, the chloride of B, aluminum, Ga, In, Tl, Si, germanium, Sn, Pb, As, Sb, and Bi, fluoride, a bromide, and an iodide mention -- having -- especially --  $\text{BCl}_3$ ,  $\text{BBr}_3$ ,  $\text{BI}_3$ ,  $\text{AlCl}_3$ ,  $\text{AlBr}_3$ ,



GaCl<sub>3</sub>, GaBr<sub>3</sub>, InCl<sub>3</sub>, TiCl<sub>3</sub>, SiCl<sub>4</sub>, SnCl<sub>4</sub>, SbCl<sub>5</sub>, and SbF<sub>5</sub> etc. -- it is suitable.

[0031] Contact to a component 1, a component 2, a component 3 and a component 4, and the halogen content compound that can be contacted further if needed is made by carrying out mixed stirring or co-grinding mechanically under existence of an inactive medium or un-existing. Contact can be performed under 40-150-degree C heating.

[0032] As an inactive medium, aromatic hydrocarbon, such as saturation alicyclic hydrocarbon, such as saturated aliphatic hydrocarbon, such as a hexane, a heptane, and an octane, a cyclopentane, and a cyclohexane, benzene, toluene, and a xylene, can use it.

[0033] As an example of the method of preparation of the component A in this invention, the approach currently indicated by JP,58-162607,A, 55-94909, 55-115405, 57-108107, 61-21109, 61-174204, 61-174205, 61-174206, the 62-7706 official report, etc. is mentioned. In a detail, it is [0034] more. \*\* The resultant of a metallic oxide and magnesium JIARUKOKISHIDO How (JP,58-162607,A) to make an electron-donative compound and tetravalent halogenation titanium contact, \*\* The resultant of an inorganic oxide and a magnesium hydrocarbyl halide compound How (JP,55-94909,A) to make a Lewis base compound and a titanium tetrachloride contact, \*\* The resultant of porosity support, such as a silica, and an alkyl magnesium compound the approach (JP,55-115405,A --) of making an electron-donative compound and a silicon halide compound contacting before making a titanium compound contacting A 57-108107 official report, a \*\* metallic oxide, an alkoxy group content magnesium compound, How (JP,61-174204,A) to contact the aromatic series multiple-valued carboxylic acid which has a carboxyl group in the ortho position or its derivative, and a titanium compound, \*\* A metallic oxide, an alkoxy group content magnesium compound, the silicon compound that has hydrogen-silicon association, How (JP,61-174205,A) to contact an electron-donative compound and a titanium compound, \*\* A metallic oxide, an alkoxy group content magnesium compound, a halogen, or a halogen content compound, How (JP,61-174206,A) to contact an electron-donative compound and a titanium compound, \*\* The resultant acquired by contacting a metallic oxide, dihydrocull BIRUMAGUNESHIUMU, and halogen content alcohol How (JP,61-21109,A) to make an electron-donative compound and a titanium compound contact, \*\* The solid-state obtained by contacting a metallic oxide, hydrocarbyl magnesium, and a hydrocarbyloxy radical content compound (equivalent to said alkoxy group content compound) It is the approach (JP,62-7706,A) of making halogen content alcohol contacting and making an electron-donative compound and a titanium compound contacting further. Especially the approach of \*\* and \*\* has the desirable approach of \*\* - \*\* also among these.

[0035] Although Component A is prepared as mentioned above, the aforementioned inactive medium may wash Component A if needed, and it may be dried further.

[0036] Moreover, Component A may make the olefin polymer which is contacted to an olefin and generated in Component A contain under existence of an organoaluminium compound further. As an organoaluminium compound, it is chosen from the after-mentioned organoaluminium compounds.

[0037] As an olefin, alpha olefins, such as other propylenes of ethylene, 1-butene, 1-hexene, and 4-methyl-1-pentene, can use it. It is desirable to perform contact to an olefin under existence of the aforementioned inactive medium. 100 degrees C or less of contact are usually desirably performed at the temperature of -10-+50 degrees C. the amount of the olefin polymer made to contain in Component A -- Component A -- it is usual 0.1-100g per g.

[0038] Contact of Component A and an olefin may make an electron-donative compound exist with an organoaluminium compound. An electron-donative compound is chosen from the organosilicon compounds which have the compound used in case Component A is prepared and Si-O-C association, or Si-N-C association. The aforementioned inactive medium can wash the component A in contact with an olefin if needed, and it can be dried further.

[0039] An organoaluminium compound organoaluminium compound (henceforth Component B) General formula  $R_{14}n_1AlX_3-n_1$  (however, in R<sub>14</sub>, an alkyl group or an aryl group, and X show a halogen atom, an alkoxy group, or a hydrogen atom, and n<sub>1</sub> is the number of the arbitration of the range of  $1 \leq n_1 \leq 3$ .) It is what is shown. For example, trialkylaluminium and dialkyl aluminum mono-halide, Monoalkyl aluminum dihalide, alkylaluminium sesquihalide, There are no carbon numbers 1,



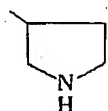
such as dialkylaluminum monoalkoxide and a dialkyl aluminum mono-hydride, and a carbon number 2 thru/or six alkylaluminum compound, its mixture, or a complex compound is especially preferably desirable 18 pieces. Specifically Trimethylaluminum, triethylaluminum, TORIPURO pill aluminum, Triisopropyl aluminum, tributyl aluminum, triisobutylaluminum, Trialkylaluminums, such as trihexyl aluminum, dimethyl aluminum chloride, A diethylaluminum chloride, diethyl aluminum bromide, diethyl aluminum iodide, Dialkyl aluminum mono-halide, such as diisobutyl aluminum chloride, Methyl aluminum jib ROMIDO methyl aluminum dichloride and ethyl aluminum dichloride -- ethyl aluminum - a jib -- ROMIDO and ethyl aluminum NIUMUJI iodide -- Monoalkyl aluminum dihalide, such as isobutyl aluminum dichloride, Alkylaluminum sesquihalide, such as ethylaluminum sesquichloride, A dimethyl aluminum methoxide, diethyl aluminum ethoxide, A diethyl aluminum phenoxide, dipropyl aluminum ethoxide, Dialkylaluminum monoalkoxides, such as diisobutyl aluminum ethoxide and a diisobutyl aluminum phenoxide, Dialkyl aluminum hydrides, such as a dimethyl aluminum hydride, a diethyl aluminum hydride, a dipropyl aluminum hydride, and a diisobutyl aluminum hydride, are mentioned. Also in these, especially triethylaluminum and triisobutylaluminum have desirable trialkylaluminum.

[0040] The organosilicon compound (henceforth Component C) which is 1 of the catalyst component used by organosilicon compound this invention component is expressed with said general formula. In this type, they are R1 and a \*\*\*\*\* atom content heterocycle type substituent. The carbon atom of the arbitration within heterocycle couples this substituent directly with the silicon atom of Component C. As this substituent, ethyleneimine, azetidine, a pyrrole, a pyrrolidine, A pyrazole, an imidazole, triazole, oxazole, furazan, A thiazole, a pyridine, HIPERIJIN, pyridazine, a pyrimidine, pyrazine, A piperazine, oxazine, a morpholine, the thiazin, Indore, indoline, Indazole, benzimidazole, benzotriazol, benzooxazole, The radical with the skeletal structure of a pudding, a quinoline, cinnoline, quinazoline, quinoxaline, phthalazine, a NAFUCHI lysine, a pteridine, a carbazole, an acridine, phenazine, etc. of monovalence is mentioned. The following and substituent R1 An example is shown. In addition, in the following, Me shows a methyl group.

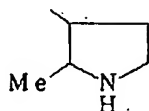
[0041]

[Formula 3]

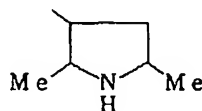
RA :



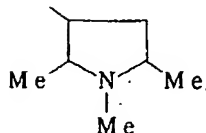
RB :



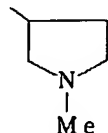
RC :



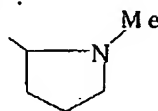
RD :



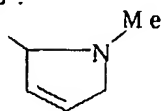
RE :



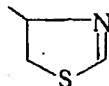
RF :



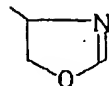
RG :



RH :



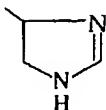
RI :



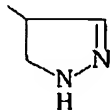
[0042]

[Formula 4]

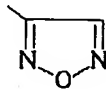
RJ :



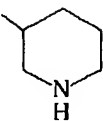
RK :



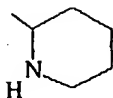
RL :



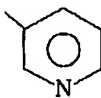
RM :



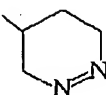
RN :



RO :



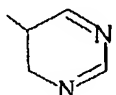
RP :



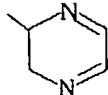
[0043]

[Formula 5]

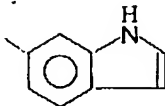
RQ :



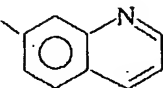
RR :



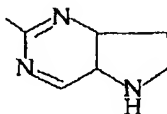
RS :



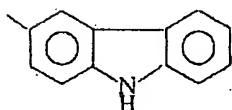
RT :



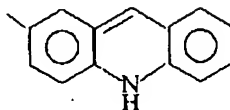
RU :



RV :



RW :



Especially the thing of five membered-rings and six membered-rings is desirable among the above-mentioned substituents.

[0044] R2 in said general formula of Component C R4 in a hydrocarbon group and R4 O-, R5 3 Si-, and R6 3 SiO-, R5, and R6 As a hydrocarbon group, an alkyl group, an alkenyl radical, a cycloalkyl radical, a cyclo alkenyl radical, a cyclo alkadienyl radical, an aryl group, an aralkyl radical, etc. are mentioned. As an alkyl group, methyl, ethyl, propyl, i-propyl, butyl, i-butyl, s-butyl, t-butyl, amyl, i-amyl, t-amyl, hexyl, octyl, 2-ethylhexyl, a decyl group, etc. as an alkenyl radical Vinyl, an allyl compound, propenyl, isopropenyl, 1-butenyl, 1-pentenyl, 1-hexenyl, 1-OKUTENIRU, 1-DEKENIRU, 1-methyl-1-cutting-pliers nil, 1-methyl-1-heptenyl, etc. as a cycloalkyl radical Cyclo butyl, cyclopentyl, cyclohexyl, a methylcyclohexyl radical, etc. as a cyclo alkenyl radical Cyclo pentenyl, cyclohexenyl, a methyl cyclohexenyl group, etc. as a cyclo alkadienyl radical Phenyl, tolyl, a xylyl group, etc. are mentioned as

an aryl group, and cyclopentadienyl, methylcyclopentadienyl, an indenyl group, etc. are mentioned for benzyl, phenethyl, 3-phenylpropyl radical, etc. as an aralkyl radical.

[0045] Hereafter, Component C is illustrated. It is R1 [ in / on the following and / in signs, such as [RD] and [RF], / the general formula of Component C ]. In Me, corresponding to the aforementioned sign, methyl and Et show ethyl and Pr shows propyl, respectively. [RD] Si (OMe)<sub>3</sub>, [RD] Si () [ OSiMe<sub>3</sub> ] (OMe)<sub>2</sub>, [RF] Si (Oi-Pr) (OMe)<sub>2</sub>, [RF] Si (OEt)<sub>3</sub>, [RG] Si (OEt)<sub>3</sub>, [RG] Si (Oi-Pr) (OMe)<sub>2</sub>, [RE] Si (OMe)<sub>3</sub>, [RE] Si () [ OSiMe<sub>3</sub> ] (OMe)<sub>2</sub>, [RP] Si (OMe)<sub>3</sub>, and [RQ] Si<sub>3</sub> (OEt).

[0046] Precuring of a precuring solid-state component (component A) is made by making an olefin (component D) contact under existence of the organosilicon compound (component C) shown by the organoaluminium compound (component B) and said general formula.

[0047] Moreover, an electron-donative compound (henceforth Component E) may also be added with Component B and Component C if needed at the time of precuring of Component A. The electron-donative compound which contains hetero atoms, such as an electron-donative compound which consists of an organosilicon compound, and nitrogen, sulfur, oxygen, Lynn, as an electron-donative compound is also usable.

[0048] Although the organosilicon compound which has Si-O-C association or Si-N-C association is mentioned as an organosilicon compound, it is the compound which has Si-O-C association desirably. as such a compound -- general formula R15n3Si(OR16)4-n3 -- [ -- however, R15 shows a hydrocarbon group or a halogen atom, and R16 shows a hydrocarbon group and 0 ≤ n3 ≤ 3. ] It comes out and the compound expressed is mentioned. As a hydrocarbon group of R15 in the above-mentioned general formula, an alkyl group, a cycloalkyl radical, an aryl group, an aralkyl radical, an alkenyl radical, an alkadienyl radical, a cyclo alkenyl radical, a SHIKUROARU rudder nil radical, etc. are mentioned. Chlorine, a bromine, iodine, etc. are mentioned as a halogen atom of R15. Moreover, as a hydrocarbon group of R16, an alkyl group, a cycloalkyl radical, an aryl group, an alkenyl radical, etc. are mentioned. n3 [ furthermore, ] The hydrocarbon group of 16 ORR16 of an individual (4-n3) may be the same as the hydrocarbon group of R15 of an individual, or may differ from each other.

[0049] As the example, a tetramethoxy silane, a tetra-ethoxy silane, Tetra-butoxysilane, tetrapod iso-butoxysilane, a tetra-phenoxy silane, A tetrapod (p-methylphenoxy) silane, a tetra-benzoyloxy silane, Methyl trimethoxysilane, methyl triethoxysilane, MECHIRUTORI butoxysilane, Methyl triphenoxysilane, ethyltriethoxysilane, ethyl TORIISO butoxysilane, Ethyl triphenoxysilane, butyltrimethoxysilane, epoxybutyltriethoxysilane, BUCHIRUTORI butoxysilane, butyl triphenoxysilane, isobutyl TORIISO butoxysilane, Vinyltriethoxysilane, allyl compound trimethoxysilane, dimethyl diisopropoxysilane, Dimethyl dibutoxysilane, a dimethyl dihexyl oxy-silane, a JIMECHIRUJI phenoxy silane, Diethyldiethoxysilane, diethyl JIISO butoxysilane, a JIECHIRUJI phenoxy silane, dibutyl diisopropoxysilane, dibutyl dibutoxysilane, and a jib -- a CHIRUJI phenoxy silane -- Diisobutyl diethoxysilane, diisobutyl JIISO butoxysilane, Diphenyldimethoxysilane, diphenyl diethoxysilane, diphenyl dibutoxysilane, Dibenzyl diethoxysilane, a JIBINIRUJI phenoxy silane, a diaryl dipropoxy silane, a diphenyl diaryl oxy-silane, methylphenyl dimethoxysilane, chlorophenyl diethoxysilane, etc. are mentioned.

[0050] As an example of the electron-donative compound containing a hetero atom As a compound containing a nitrogen atom, 2, 2, 6, and 6-tetramethylpiperidine, 2, 6-dimethyl piperidine, 2, 6-diethyl piperidine, 2, 6-diisopropyl piperidine, 2, a 6-diisobutyl-4-methyl piperidine, 1, 2, 2 and 6, 6-pentamethylpiperidine, A 2, 2, 5, and 5-tetramethyl pyrrolidine, 2, 5-dimethyl pyrrolidine, 2, 5-diethyl pyrrolidine, 2, 5-diisopropyl pyrrolidine, 1, 2, 2, 5, and 5-pentamethyl pyrrolidine, 2 and 2, a 5-trimethyl pyrrolidine, 2-methylpyridine, 3-methylpyridine, 4-methylpyridine, 2, 6-diisopropyl pyridine, 2, a 6-diisobutyl pyridine, 1 and 2, a 4-trimethyl piperidine, 2, 5-dimethyl piperidine, the methyl nicotinate, nicotinic-acid ethyl, Nicotinamide, a benzoic-acid amide, 2-methyl pyrrole, 2, 5-dimethyl pyrrole, An imidazole, a toluic-acid amide, a benzonitrile, an acetonitrile, An aniline, p-toluidine, an alt.toluidine, a meta-toluidine, triethylamine, diethylamine, dibutyl amine, a tetramethylenediamine, tributylamine, etc. as a compound containing a sulfur atom A thiophenol, a thiophene, 2-thiophene carboxylic-acid ethyl, 3-thiophene carboxylic-acid ethyl, 2-methylthiophene, methyl mercaptan, ethyl mercaptan, An isopropyl

mercaptan, butyl mercaptan, a diethyl thioether, a diphenyl thioether, benzenesulfonic acid methyl, a methyl ape fight, an ethyl ape fight, etc. as a compound containing an oxygen atom A tetrahydrofuran, 2-methyl tetrahydrofuran, 3-methyl tetrahydrofuran, 2-ethyl tetrahydrofuran, 2, 2 and 5, 5-tetraethyl tetrahydrofuran, A 2, 2, 5, and 5-tetramethyl tetrahydrofuran, 2, 2 and 6, 6-tetraethyl tetrahydropyran, 2, 2, 6, and 6-tetramethyl tetrahydropyran, dioxane, wood ether; Diethylether, dibutyl ether, JISO amyl ether, diphenyl ether, An anisole, an acetophenone, an acetone, a methyl ethyl ketone, an acetylacetone, An o-tolyl-t-butyl ketone, methyl -2, a 6-JI t-buthylphenyl ketone, 2-FURARU acid ethyl, 2-FURARU acid isoamyl, 2-FURARU acid methyl, 2-FURARU acid propyl, etc. as a compound containing the Lynn atom Triphenyl phosphine, tributyl phosphine, triphenyl phosphite, tribenzyl phosphite, diethyl phosphate, diphenyl phosphate, etc. are mentioned.

[0051] Two or more sorts of these electron-donative compounds may be used. Moreover, in case an organometallic compound is used for these electron-donative compound combining a catalyst component, it may be used, and after making an organometallic compound contact beforehand, it may be used.

[0052] As an olefin, alpha olefins, such as a propylene besides ethylene, 1-butene, 1-hexene, and 4-methyl-1-pentene, can use it. As for precuring, it is desirable to carry out under existence of the aforementioned inactive medium. precuring -- usually -- the temperature of 100 degrees C or less -30 degrees C - +50 degrees C are desirably performed still more desirably at the temperature of -20 degrees C - +15 degrees C. As a polymerization method, any of a batch type and continuous system are sufficient, and you may carry out multistage [ two or more steps of ]. When carrying out multistage, naturally polymerization conditions can be changed, respectively.

[0053] Component B -- the concentration in a precuring system -- 10 - 500 millimol / liter -- it is used so that it may become 30 - 200 millimol / liter desirably, and 1-50,000 mols per titanium 1 gram atom in Component A are used so that it may become 2-50 mols desirably.

[0054] Component C -- the concentration in a precuring system -- 5 - 1000 millimol / liter -- it is used so that it may become 10 - 200 millimol / liter desirably.

[0055] although an olefin polymer is incorporated in Component A by precuring -- the amount of polymers -- Component A -- it is desirable to be referred to especially as 0.5-50g 0.1-200g perg.

[0056] the electron-donative compound used if needed -- the concentration in a precuring system -- 1 - 100 millimol / liter -- it is used so that it may become 5 - 50 millimol / liter desirably.

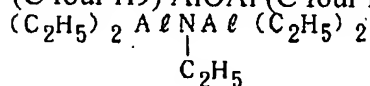
[0057] Although the aforementioned inactive medium can dilute or wash the catalyst component obtained as mentioned above, it is desirable to wash especially from a viewpoint which prevents preservation degradation of a catalyst component. You may dry after washing if needed. Moreover, when saving a catalyst component, saving at the made length low temperature is desirable, and -50 degrees C - +30 degrees C of temperature requirements which are -20 degrees C - +5 degrees C are recommended especially.

[0058] The catalyst component which is the polymerization above of an alpha olefin, and was made and obtained An organometallic compound and also if needed, combining an electron-donative compound, although it is useful as a catalyst of copolymerization with homopolymerization of the alpha olefin of 3-10 carbon numbers, other monoolefins, or the diolefin of 3-10 carbon numbers The engine performance which was extremely excellent especially as homopolymerization, such as a carbon number 3 thru/or six alpha olefins, for example, a propylene, 1-butene, 4-methyl-1-pentene, and 1-hexene, or both above alpha olefins and/or random with ethylene, and a catalyst of block copolymerization is shown.

[0059] the organometallic compound which can be used -- the [ the Ith group of a periodic table thru/or ] -- III It is the organic compound of a group metal. As this compound, a lithium, magnesium, calcium, zinc, and the organic compound of aluminum can use it. An organoaluminium compound is suitable also especially in these. Although suitably chosen as an organoaluminium compound which can be used from the compounds used in the case of precuring of said solid-state catalyst component (component A), trialkylaluminium especially triethylaluminum, and triisobutylaluminum are desirable. Moreover, these trialkylaluminums can be used together with other organoaluminium compounds, for example, the diethylaluminium chloride which is easy to come to hand industrially, ethyl aluminum dichloride,

ethylaluminium sesquichloride, diethyl aluminum ethoxide, diethyl aluminum hydrides, such mixture, or a complex compound.

[0060] Moreover, the organoaluminium compound which two or more aluminum combined through the oxygen atom or the nitrogen atom is also usable. As such a compound, they are  $2 \text{ AlOAl} (\text{C four H9})_2$ ,  $2 (\text{C four H9}) \text{ AlOAl} (\text{C four H9})_2$ , and [Formula 6], for example (C two H5).



\*\* can be illustrated.

[0061] as the organic compound of metals other than an aluminum metal -- other  $\text{LiAl(s)} (\text{C two H5})_4$ , such as diethyl magnesium, ethyl magnesium chloride, and diethylzinc, and  $\text{LiAl} (\text{C seven H15})_4$  etc. -- a compound is mentioned.

[0062] As an electron-donative compound combinable if needed [ above / catalyst component and organometallic compound, and if needed ], it is suitably chosen from the electron-donative compounds which may be used in the case of precuring of said component A. Two or more sorts of these electron-donative compounds may be used. Moreover, in case an organometallic compound is used for these electron-donative compound combining a catalyst component, it may be used, and after making an organometallic compound contact beforehand, it may be used.

[0063] Especially the amount of the organometallic compound used to the aforementioned catalyst component usually has 20 to 500 desirable gram mol one to 2,000 gram mol per titanium 1 gram atom in this catalyst.

[0064] moreover, the ratio of an organometallic compound and an electron-donative compound -- one mol of electron-donative compounds -- receiving -- an organometallic compound -- as aluminum -- 0.1-40 -- it is preferably chosen in the range of 1 - 25 gram atom.

[0065] Copolymerization of the copolymerization propylene of a propylene is performed by copolymerizing a propylene and ethylene under existence of the aforementioned polymerization catalyst.

[0066] Any of a gaseous phase and the liquid phase are sufficient as a polymerization reaction, and when carrying out a polymerization by the liquid phase, it can be performed in inert hydrocarbon, such as normal butane, an isobutane, a normal pentane, an isopentane, a hexane, a heptane, an octane, a cyclohexane, benzene, toluene, and a xylene, and a liquefied monomer. The range of -80 degrees C - +150 degrees C of polymerization temperature is usually 40-120 degrees C preferably. The polymerization preassure force is good with for example, one to 60 atmospheric pressure.

[0067] The operating rate of a propylene and ethylene is suitably set up so that the ethylene content in the copolymer obtained may become 0.1 - 5 % of the weight desirably 0.01 to 10% of the weight. Moreover, it is desirable for a copolymerization reaction to make [ 0.01-200g / ] 0.1-100g / 10 minutes especially for 10 minutes the melt flow rate (MFR) of the copolymer obtained, and the accommodation is made by usually adjusting the amount of hydrogen or the well-known molecular weight modifier used.

[0068]

[Example] An example and the example of a comparison explain this invention concretely. In addition, especially the percent (%) in an example is based on weight, unless it refuses. Physical-properties measurement of a polymer BHT (2, 6-JITA challis butyl-4-methyl phenol) to this polymer powder 0.18 % of the weight, DSTDP (distearyl thiodipropionate) 0.08 % of the weight, IRGANOX1010 [tetrakis-{methylene-(3, 5-JITA challis butyl-4-hydroxy hydronalium-cinnamate) methane}] 0.04 % of the weight, After adding calcium stearate 0.06% of the weight, respectively and considering as a pellet by melting kneading, it carried out by creating a test piece with injection molding (it considered as the sheet with a thickness of 1.0mm at measurement of Hayes). bending elastic-modulus: -- JIS K 7203-1982 -- :Hayes: -- ASTM D1003 -- conformity. Moreover, MFR of a polymer is ASTM. It measured according to D-1238. The ethylene content of a copolymer measured and asked for the permeability of absorption of 733cm-1 with the infrared absorption spectral method. In addition, the test piece carried out annealing

processing and obtained what was used as the film with a thickness of about 0.5mm by press forming. [0069] Nitrogen gas permuted the 200ml flask furnished with the preparation dropping funnel and agitator of the example 1 component A. 5g and 40ml of n-heptanes were put into this flask for what calcinated silicon oxide (the product made from DAVISON, trade name G-952) at 700 more degrees C by 200 degrees C in the nitrogen air current for 5 hours for 2 hours. Furthermore 20ml (the product made from Texas ARUKIRUZU, trade name MAGALA BEM) of 20% n-heptane solutions of n-butyl ethyl magnesium (henceforth BEM) was added, and it stirred at 90 degrees C for 1 hour. After cooling the above-mentioned suspension at 0 degree C, the solution which dissolved tetra-ethoxy silane 11.2g in 20ml n-heptane was dropped at this over 30 minutes from the dropping funnel. After dropping termination, the temperature up was carried out to 50 degrees C over 2 hours, and stirring was continued at 50 degrees C for 1 hour. The decantation removed supernatant liquor after reaction termination, 60ml n-heptane washed the generated solid-state at the room temperature, and the decantation removed supernatant liquor further. Washing processing by this n-heptane was performed further 4 times. 50ml n-heptane was added and it considered as suspension, and from the dropping funnel, in 25 degrees C, the solution which dissolved 2, 2, and 2-trichloroethanol 8.0g in 10ml n-heptane at this was hung on the above-mentioned solid-state for 15 minutes, and was dropped at it. Stirring was continued for 30 minutes at 25 degrees C after dropping termination. In the room temperature, 60ml toluene washed 3 times twice in 60ml n-heptane after reaction termination, respectively. When the obtained solid-state (solid-state component I) was analyzed, 38.5% of chlorine was included SiO<sub>2</sub> 36.6% and magnesium 5.1%. n-heptane 10ml and 40ml of titanium tetrachlorides were added to the solid-state component I obtained above, and the temperature up was carried out to 90 degrees C, and 0.6g of di-n-butyl phthalate which dissolved in n-heptane 5ml was hung for 5 minutes, and it added. Then, the temperature up was carried out to 115 degrees C, and it was made to react for 2 hours. After lowering the temperature at 90 degrees C, the decantation performed washing twice by n-heptane 70ml except for supernatant liquor. Furthermore, n-heptane 15ml and 40ml of titanium tetrachlorides were added, and it was made to react at 115 degrees C for 2 hours. The obtained solid matter was performed in 60ml n-hexane after reaction termination, and washing was performed 8 times at the room temperature. Subsequently, desiccation was performed at the bottom room temperature of reduced pressure for 1 hour, and the 8.3g catalyst component (component A) was obtained. The other silicon oxide, chlorine, and di-n-butyl phthalate of 3.1% of titanium were contained in this component A.

[0070] It cooled at 0 degree C, putting in and stirring component A 3.1g and n-heptane 300ml obtained by the above under nitrogen-gas-atmosphere mind to the 500ml reactor furnished with a precuring agitator. Next, it added so that the concentration of TEAL in the system of reaction and 2 and 3, and 4-trimethyl-3-azacyclo pentyl trimethoxysilane might serve as 80 millimols / liter, and 10 millimols / liter, respectively, and n-heptane solution (2.0 mols/(l.)) of triethylaluminum (it calls for short Following TEAL.) and 2 and 3, and 4-trimethyl-3-azacyclo pentyl trimethoxysilane were stirred for 5 minutes. Subsequently, after decompressing the inside of a system, propylene gas was supplied continuously and the polymerization of the propylene was carried out for 4.0 hours. The propylene of a gaseous phase was purged with nitrogen gas after polymerization termination, it washed each by 100ml n-heptane, and the solid phase section was washed at the room temperature 3 times. Furthermore, reduced pressure drying of the solid phase section was carried out at the room temperature for 1 hour, and the catalyst component was prepared. the result of having measured the amount of magnesium contained in a catalyst component -- the amount of precuring -- Component A -- it was 2.7g perg.

[0071] In precuring of two to example 4 example 1 instead of 2, 3, and 4-trimethyl-3-azacyclo pentyl trimethoxysilane About the silane compound shown in Table 1, they are TEAL or triisobutylaluminum (it calls for short Following TIBAL.) again. Except having carried out as it used by the concentration shown in Table 1, respectively, and it used by the concentration which shows the electron-donative compound shown in Table 1 in Table 1, respectively and precuring conditions were shown in Table 1, precuring of Component A was performed like the example 1, and the catalyst component was prepared.

[0072] In example of comparison 1 example 1, the catalyst component was prepared like the example 1

except having not performed precuring.

[0073] In precuring of example of comparison 2 example 1, except having carried out not using 2, 3, and 4-trimethyl-3-azacyclo pentyl trimethoxysilane, as precuring conditions were shown in Table 1, precuring of Component A was performed like the example 1, and the catalyst component was prepared.

[0074] In precuring of example of comparison 3 example 1, except having carried out as it used by the concentration which shows TEAL and diphenyldimethoxysilane in Table 1 not using 2, 3, and 4-trimethyl-3-azacyclo pentyl trimethoxysilane and precuring conditions were shown in Table 1, precuring of Component A was performed like the example 1, and the catalyst component was prepared.

[0075]

[Table 1]



表 1

例	シラン化合物		有機アルミニウム		電子供与性化合物		予備重合		
	名称	濃度 ミリモル/リットル	名称	濃度 ミリモル/リットル	名称	濃度 ミリモル/リットル	温度 (°C)	時間 (h)	重合量 (g/g)
実施例 1	[RD]Si(OMe) <sub>3</sub>	10	TEAL	80	—	—	0	4.0	2.7
" 2	[RD]Si(OEt) <sub>3</sub>	10	TEAL	100	PhCOEt	5	5	4.5	2.0
" 3	[RF]Si(Oi-Pr)(OMe) <sub>2</sub>	20	TEAL	80	—	—	-5	4.0	2.2
" 4	[RG]Si(OEt) <sub>3</sub>	10	TIBAL	60	—	—	0	3.0	2.5
比較例 1	予備重合なし								
" 2	—	—	TEAL	100	—	—	0	3.0	7.5
" 3	—	—	TEAL	80	Ph <sub>2</sub> Si(OMe) <sub>2</sub>	10	5	2.0	5.3

[0076] 100mg of polymerization catalyst components obtained above, 4ml of solutions which contain 0.3-mol triethyl aluminum UNIMU in 1l. of n-heptanes, and 3ml of solutions which contain 0.08-mol diphenyldimethoxysilane in 1l. of n-heptanes were mixed to the 5l. autoclave which carried out the copolymerization nitrogen purge of a propylene and was fully dried, and what was held for 5 minutes was put into it. Subsequently, after pressing 2.5l. and 3l. of liquid propylenes fit for hydrogen, the temperature up of the interior temperature of an autoclave was carried out to 70 degrees C, ethylene was supplied continuously, and the polymerization was performed for 1 hour. An unreacted propylene,

ethylene, and hydrogen were purged after polymerization termination, and the polymer in an autoclave was taken out. MFRs of the obtained polymer were 15.7g / 10 minutes, and the ethylene content was 2.8%. When physical properties were measured,  $10.79 \times 10^3$  kg/cm<sup>2</sup> and Hayes of the bending elastic modulus were 19.3%. Those results were shown in Table 2.

[0077] Using the polymerization catalyst component obtained in examples 2-4 and the examples 1-3 of a comparison, copolymerization of a propylene was performed like the above and those results were shown in Table 2. Although the bending elastic modulus of a copolymer and the value of Hayes which were acquired in each example and the example of a comparison were plotted to drawing 1, as for the copolymer obtained in each example, it is clear from the result of drawing 1 that a bending elastic modulus and the physical-properties balance of Hayes are excellent compared with them of the copolymer obtained in the example of a comparison.

[0078]

[Table 2]

表 - 2

	M F R	エチレン含量	曲げ弾性率	ヘイズ
	(g / 10分)	(%)	( $10^3$ kg / cm <sup>2</sup> )	(%)
実施例 1	15.7	2.8	10.79	19.3
" 2	17.1	2.3	11.02	21.2
" 3	16.2	2.9	10.61	18.7
" 4	15.9	1.9	11.30	23.1
比較例 1	15.8	2.4	10.48	21.2
" 2	15.6	2.9	10.13	19.1
" 3	16.0	2.0	10.72	22.6

[0079]

[Effect of the Invention] By enforcing the approach of this invention, the polypropylene copolymer excellent in the balance of transparency and rigidity can be obtained.

[Translation done.]